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PATENT

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St. Louis, Missouri
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CERTIFICATE

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In re application of:	:	
Gregory M. Jones et al.	:	
	:	
Serial No.: 09/509,126	:	Examiner M. Cygan
	:	
Filed: March 22, 2000	:	Group Art Unit 2856
	:	
For: MEASURMENT AND CONTROL OF ASPHALTENE AGGLOMERATION IN HYDROCARBON LIQUIDS	:	

Board of Patent Appeals & Interference
Room 10C01, Crystal Gateway 2
1225 Jefferson Davis Highway
Arlington, Virginia 22202

APPEAL BRIEF

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(1) Real party in Interest:

The real parties in interest are Baker Hughes Corporation and The University of Leeds, by virtue of assignments of the entire right and interest from the named inventors.

(2) Related Appeals and Interferences:

There are no pending Appeals or Interferences.

(3) Status of Claims:

Claims 1-26 are pending. Claims 1-22 and 24-26 have been rejected. Claim 23 has been objected to. Applicants appeal from the rejections of claims 1-22 and 24-26 and from the objection to claim 23.

(4) Status of Amendments:

The only response filed after final rejection, Response B filed by Certificate of Mailing dated July 3, 2001, contained no amendments.

(5) Summary of the Invention:

As defined by claim 1, the present invention is directed to a method for measuring the agglomerative state of asphaltenes in oil containing asphaltenes. The method comprises applying to the oil a series of pulses of acoustic energy. Each pulse comprises acoustic energy at multiple frequencies. At least a part of the energy is thereby scattered. For each of a plurality of pulses in the series, the scattered acoustic energy is detected to produce amplitude versus time data. The amplitude versus time data is resolved to obtain a magnitude of the detected scattered acoustic energy at selected frequencies. The magnitude for each pulse at each selected frequency is averaged over the plurality of pulses. The agglomerative state of the asphaltenes is then determined from the averaging. See, for example, Spec.[1] p. 6, lines 9-17; Spec. p. 16, line 4 - p. 21, line 9; and Spec. Fig. 4.

As defined by claim 2, the present invention is directed to a method as set forth in claim 1 wherein the selected frequencies comprise at least three different frequencies. See, for example, Spec. p. 18, lines 5-7.

[1]As used herein, "Spec." refers to the subject specification.

As defined by claim 3, the present invention is directed to a method as set forth in claim 1 wherein the selected frequencies comprise at least fifteen different frequencies. See, for example, Spec. p. 18, lines 7-10.

As defined in claim 4, the present invention is directed to a method as set forth in claim 1 wherein the steps are carried out without diluting the hydrocarbon liquid. See, for example, Spec. p. 6, lines 18-20; and Spec. p. 14, lines 10-15.

As defined in claim 5, the present invention is directed to a method as set forth in claim 4 wherein the steps of the method are carried out substantially instantaneously. See, for example, Spec. p. 18, line 31 - p. 19, line 4.

As defined in claim 6, the present invention is directed to a method as set forth in claim 5, wherein the detected scattered acoustic energy is back-scattered acoustic energy. See, for example, Spec. p. 12, lines 10-14.

As defined in claim 7, the present invention is directed to a method as set forth in claim 6, wherein the scattered acoustic energy is detected over a frequency range of from about 0.1 MHz to about 20 MHz. See, for example, Spec. p. 16, lines 4-6; and Spec. p. 22, line 32 - p. 23, line 2.

As defined in claim 8, the present invention is directed to a method as set forth in claim 7, wherein the scattered acoustic energy is detected over a frequency range of from about 0.1 MHz to about 200 MHz. See, for example, Spec. p. 15, lines 2-4; and Spec. p. 16, lines 8-9.

As defined in claim 9, the present invention is directed to a method as set forth in claim 8, wherein the scattered acoustic energy is detected over a frequency range of from about 14 MHz to about 20 MHz. See, for example, Spec. p. 18, lines 23-25 and 28-30.

As defined in claim 10, the present invention is directed to a method as set forth in claim 1, wherein the detecting is carried out by at least one sensor, which sensor is incorporated in a signal input probe. See, for example, Spec. p. 10, lines 26-28.

As defined in claim 11, the present invention is directed to a method as set forth in claim 1, wherein the detecting is carried out by at least one sensor which sensor is separate from a signal input probe. See, for example, Spec. p. 10, lines 28-29.

As defined in claim 12, the present invention is directed to a method as set forth in claim 11, wherein the signal input probe and the sensor are located so that the signal direction of the probe intersects the signal direction of the sensor at an angle of less than about 90°. See, for example, Spec. p. 11, lines 4-8 and 25-29.

As defined in claim 13, the present invention is directed to a method as set forth in claim 12, wherein the signal input probe and the sensor are located so that the signal direction of the probe intersects the signal direction of the sensor at an angle of less than about 60°. See, for example, Spec. p. 11, lines 25-29.

As defined in claim 14, the present invention is directed to a method as set forth in claim 13, wherein the signal input probe and the sensor are located so that the signal direction of the probe intersects the signal direction of the sensor at an angle of less than about 45°. See, for example, Spec. p. 11, lines 25-29.

As defined in claim 15, the present invention is directed to a method as set forth in claim 1, wherein the resolving of the amplitude versus time data comprises gating the detected scattered acoustic energy to that part of the detected energy emanating from a focal region and Fourier transforming the amplitude versus time data into a magnitude vs. frequency format. See, for example, Spec. p. 17, lines 9-14.

As defined in claim 16, the present invention is directed to a method as set forth in claim 1, wherein the pulses of acoustic energy are applied as a tone-burst and the step of resolving of the amplitude versus time data comprises detecting the magnitude of the scattered energy at selected frequencies. See, for example, Spec. p. 9, lines 24-26.

As defined in claim 17, the present invention is directed to a method as set forth in claim 1, wherein the averaging over the series of pulses the magnitude for each pulse at each selected frequency produces an average of the magnitude for each selected frequency, and the determining of the agglomerative state of the asphaltenes is effected by comparing the that average for each selected frequency with a standard. See, for example, Spec. p. 6, line 18- p. 7, line 2.

As defined in claim 18, the present invention is directed to a method as set forth in claim 17, wherein the standard is a sample of known particle size. See, for example, Spec. p. 17, lines 15-22.

As defined in claim 19, the present invention is directed to a method as set forth in claim 17, wherein the standard is a model of particle size based on scattering theory. See, for example, Spec. p. 17, lines 15-22.

As defined in claim 20, the present invention is directed to a method as set forth in claim 1, wherein the oil containing asphaltenes is in a process flow stream and the signal of acoustic

energy is applied to the oil in the process flow stream. See, for example, Spec. p. 14, lines 10-28.

As defined in claim 21, the present invention is directed to a method for measuring the agglomerative state of asphaltenes in an oil containing asphaltenes comprising:

a. removing a sample of the oil and without diluting the oil;

b. applying to the sample a series of pulses of acoustic energy, each pulse comprising acoustic energy at multiple frequencies, thereby scattering at least part of the energy;

c. detecting, for each of a plurality of pulses in the series, the magnitude of the scattered acoustic energy at selected frequencies to produce amplitude versus time data;

d. resolving the amplitude versus time data to obtain a magnitude of the detected scattered acoustic energy at selected incremental frequencies;

e. averaging over the plurality of pulses the magnitude for each pulse at each selected frequency;

f. deriving from the averaging a distribution of the relative size of asphaltene particles scattering acoustic energy; and

g. determining the agglomerative state of the asphaltene particles. See, for example, Spec. p. 6, line 18 - p. 7, line 2.

As defined in claim 22, the present invention is directed to a method as in claim 20, having the additional step of returning the undiluted oil sample. See for example, Spec. p. 19, lines 10-22.

As defined in claim 23, the present invention is directed to a method as in claim 20, wherein the method is carried out in a bench-scale device. See, for example, Spec. p. 10, line 32 - p. 11, line 2.

As defined in claim 24, the present invention is directed to a method for controlling the agglomeration of asphaltenes in oil which comprises applying a series of pulses of acoustic energy to the oil, each pulse comprising acoustic energy at multiple frequencies, thereby scattering at least a part of the energy; detecting, for each of a plurality of pulses in the series, the scattered energy at selected frequencies to produce amplitude versus time data; resolving the amplitude versus time data to obtain a magnitude of the detected scattered energy at selected incremental frequencies; averaging over the plurality of pulses the magnitude for each pulse at each selected frequency to obtain average magnitude versus frequency data; comparing the average magnitude versus

frequency data with a standard; and acting to control the number of particles having a particle size corresponding to the selected incremental frequencies. See, for example, Spec. p. 6, line 27 - p. 7, line 2.

As defined in claim 25, the present invention is directed to a method as set forth in claim 24, wherein the selected frequencies are limited to a frequency range of acoustic energy scattered by the agglomerated asphaltene particles characteristic of the oil. See, for example, Spec. p. 15, line 24 - p. 16, line 3.

As defined in claim 26, the present invention is directed to a method as set forth in claim 24, wherein the scattered acoustic energy is detected over a frequency range of from about 14 MHz to about 20 MHz. See, for example, Spec. p. 18, lines 23-25 and 28-30.

(6) Issues:

a. Whether claims 1-19 are unpatentable under 35 U.S.C. §103(a) as obvious over de Boer et al. SPE reference ("de Boer") in view of U.S. patent 5,853,994 to Gopinathan et al. ("the Gopinathan patent").

b. Whether claims 1-11, 15-22 and 24-26 are unpatentable under the judicially created doctrine of obviousness-type double patenting over U.S. patent 5,969,237 ("the '237 patent").

c. Whether claim 23 is still objectionable if the claim from which it depends is found to be patentable.

(7) Grouping of Claims:

The claims do not stand or fall together. Claim 23 has been objected to only and so is grouped separately from all other claims. Claims 1 - 4, 10 and 11 are directed to a method for measuring the agglomerative state of asphaltenes in oil and stand or fall together. Claims 5 and 6 require the steps of the method to be carried out substantially instantaneously and so are grouped together, but separately from claims 1 - 4, 10 and 11. Claims 7-9 identify the preferred

frequency ranges found to be particularly suitable to the present invention and claims 12-14 identify ranges of angles between probe and signal direction and so stand separately. Claim 15 calls for the resolving of the amplitude versus time data to comprise gating the detected scattered acoustic energy to that part of the detected energy emanating from a focal region and Fourier transforming the amplitude versus time data into a magnitude vs. frequency format and so stands separately. Likewise, the added features of claim 16, which calls for the pulses of acoustic energy to be applied as a tone-burst and the step of resolving of the amplitude versus time data to comprise detecting the magnitude of the scattered energy at selected frequencies, claim 17, which calls for the averaging over the series of pulses the magnitude for each pulse at each selected frequency to produce an average of the magnitude for each selected frequency, and the determination of the agglomerative state of the asphaltenes to be effected by comparing the that average for each selected frequency with a standard, claim 18, which calls for the feature noted with respect to claim 17 but wherein the standard is a sample of known particle size, and claim 19, which calls for the additional features of claim 18 wherein the standard is a model of particle size based on scattering theory, each add features as noted that nowhere seen in the art of record, yet each of such features is an important aspect of the present invention. Claims 20-22 and 24-26 are not rejected over the de Boer and Gopinathan references and so stand separately from Claims 1-19. Claim 23 stands alone as not being the subject of any rejection. Thus, the claims can be grouped as follows:

(a) Claims 1-4, 10, 11
(b) Claims 5, 6
(c) Claims 7-9
(d) Claims 12-14
(e) Claim 15
(f) Claim 16
(g) Claims 17-19
(h) Claims 20-22, 24-26

(i) Claim 23

(8) Argument and Authorities:

Reversal of the rejection of claims 1-19 as being obvious over de Boer in view of the Gopinathan patent is respectfully requested. Claim 1 (and so also claims 2-19, which depend therefrom) calls for a particular method for measuring the agglomerative state of asphaltenes in oil containing asphaltenes. The method comprises applying to the oil a series of pulses of acoustic energy. Each pulse comprises acoustic energy over a range of frequencies. At least a part of the energy is thereby scattered. The scattered acoustic energy is detected for each of a plurality of pulses in the series to produce amplitude versus time data. The amplitude versus time data is then resolved to obtain a magnitude of the detected scattered acoustic energy at selected frequencies. The magnitude for each pulse at each selected frequency is then averaged over the plurality of pulses and the agglomerative state of the asphaltenes is determined from the averaging. In short, therefore, claim 1 (and so claims 2-19) calls for applying a series of pulses of a range of frequencies of acoustic energy to the oil, detecting the resulting scattered energy over a range of frequencies for each of a plurality of the pulses to produce amplitude versus time data, which are then not averaged, but first resolved to obtain magnitude versus frequency data, which are then averaged. The agglomerative state is then determined from that averaging.

By contrast, the de Boer method involves simply receiving the back-scattering of short bursts of acoustic energy and converting that back-scattering into a read-out signal that is sorted by a multi-channel analyzer into pre-selected amplitude classes corresponding to particle sizes. For clarity, attention is directed to the Comparative Flowchart of the Methods of Claim 1 and de Boer, attached as Appendix B, which illustrates succinctly how very different the methods are. Some of the stark differences in the methods are shown perhaps more conceptually in the Comparative Schematic of the Methods of Claim 1 and de Boer, attached as Appendix C. There, the first line of the schematic for claim 1 shows a series of pulses, each illustrated by a box in which are noted the various frequencies of energy (f_1, f_2, f_3, . . .) in that pulse. For each pulse, amplitude versus time data are developed, which are then resolved to produce, for each pulse, a series of magnitudes for each frequency. The magnitudes are then not averaged together, but rather, for each frequency, the magnitudes are averaged across the pulses, to produce a series of average magnitudes for each frequency, from which the agglomerative state is determined.

By contrast, de Boer requires first pre-selecting a finite number of discrete particle sizes and relating each such size to an amplitude class. After that, de Boer describes simply applying a series of energy pulses, sorting the amplitudes into classes corresponding to the pre-determined classes, and accordingly identifying the particle size that corresponds to each class. Thus, the de Boer method is a simple sorting of amplitude into discrete, pre-selected classes. De Boer does not disclose or suggest any of the following, each of which is called for in claim 1 and so also claims 2-19:

(a) applying a series of pulses, each of which covers a range of frequencies of acoustic energy.

(b) producing amplitude versus time data.

(c) resolving the amplitude versus time data to obtain magnitude versus frequency data.

(d) averaging the magnitude versus frequency data.

(e) determining the agglomerative state from that averaging.

Instead, de Boer uses a multi-channel analyzer for separation into amplitude classes corresponding to particle sizes. Thus, while the subject claimed method measures any size particles and any number of different size particles, none of which is known prior to the measurement, and does not rely on a pre-determined correspondence between amplitude and particle size, the de Boer technique requires the pre-supposition of certain relative particle sizes and relationship between amplitude and particle sizes. Accordingly, the de Boer method is very different from that of the present claims.

Gopinathan nowhere makes up for the deficiencies of the de Boer disclosure. Gopinanthan describes a method and apparatus very different from that disclosed by de Boer and that of the present application and is applicable to neither the de Boer situation nor the present situation. But, as with de Boer and in distinction to the method of the instant claims, the Gopinathan method is dependent on a pre-selected limited number of discrete particle sizes. Thus, neither Gopinathan nor de Boer discloses or suggests a method with the flexibility to measure particle sizes and distributions thereof unencumbered with the severe limitations of pre-selecting a limited number of discrete particle sizes.

The Gopinathan method, however, is even more limited than the limited de Boer method. The Gopinathan method does not involve simple application of energy to a fluid for measurement of the sizes of particles therein. According to the Gopinathan method, up to four analytes in a liquid

may be studied by adding to the liquid a class of spherical particles for each analyte. The spherical particles of each class have a pre-determined, unique and narrow range of particle diameters and are coated with a reagent that reacts with the analyte corresponding to that class to form a conjugate formed entirely out of particles from that class (i.e., the conjugates corresponding to any one class are formed of particles of the same size). However, the difference in size between classes is significant, such as a ratio of diameters of at least 2:1. The measurements, therefore, are based on not only on pre-determined particle size classes, but also on the particles being created. The Gopinathan method is so confining that the particles to be analyzed are not native to the liquid, but must be configured artificially with the special reactant coated spheres. Thus, in Gopinathan, the particle scatterings are based on the spheres added to the liquid sample rather than the analytes themselves. And the measurements are based on each of these pre-determined and created particles having a cross-section unique to its class in distinction to the other classes of significantly different size. Accordingly, the Gopinathan method also is limited to measurements of a limited number of particle classes, each of which has a narrow range of sizes within the class and significantly different sizes between classes. (All of the preceding statements in this paragraph are supported, for example, at Gopinathan Col. 3, lines 7-21 and 29-37 and repeatedly throughout the Detailed Description of the Drawings).

The Gopinathan method, therefore, is a limited technique for assaying a limited number of analytes --generally up to four analytes-- in a liquid sample. Moreover, the Gopinathan assay appears to be inapplicable to methods for measuring the agglomerative state of asphaltenes in oil as called for in the subject claims. Gopinathan contemplates that the assay would be biological and that the liquid would be a bodily fluid. Gopinathan employs a reagent including a first class of particles that are dimensioned within a predeterminedly narrow range of particle diameters and are coated with a reactant capable of entering into a reaction involving the analyte so as to generate particles of different dimensions than those of the original classes and of characteristic acoustic scattering cross sections, and a second reagent including a second class of particles that are substantially larger than those of the first class and are coated with the noted reactant. Gopinathan forms a mixture of the reactant coated particles of the two classes with the sample to be tested so as to cause a specific reaction involving the reactant and the analyte to form conjugates of the first and second classes of particles.

Thus, in contrast to the subject claimed method and the de Boer technique, the Gopinathan

assay requires contamination of the liquid being assayed by addition of spherical particles and reactants to the liquid and inducing a reaction therein and so is carried out on a sample of the liquid rather than in-line. And, further in contrast to the subject claimed method and the de Boer technique, Gopinathan does not measure acoustic scattering by the particles but rather by conjugates formed in the reaction. The conjugates have characteristic acoustic scattering cross-sections and irradiating them with compressional waves in a predetermined frequency range causes a reduction in forward energy of the waves, which is then detected.

Clearly, therefore, the Gopinathan technique is completely different from either the de Boer method or the present method. Gopinanthan does not measure the particles directly or determine particle sizes per se, but rather reacts the particles with two reactants, measures the acoustic characteristics of the reaction products and segregates reaction product data into pre-determined categories.

Gopinathan, is simply inapplicable to situations, such as those in de Boer and the present invention, where uncontrolled agglomeration results in no set number of particle diameters nor any predetermined or set particle sizes and where addition of reactants to the tested fluid is not appropriate. And, contrary to the Examiner's assertion, the Gopinathan method is not simply a replacement of measuring magnitude versus frequency instead of amplitude versus time in de Boer. The Gopinathan is much more complicated, indirect and limited.

Accordingly, it is not seen how a method such as described by Gopinathan, wherein a limited number of discrete particle sizes must be not only present but known in advance, can be adapted to either de Boer or the present situation wherein a continuum of particles sizes of unpredictable sizes may be present. Moreover, it is not seen how the Gopinathan method, wherein scattering not by the particles but by products enlarged by two separate reactions is measured, can be applied to the de Boer and present methods where scattering by the particles themselves are measured and reactions with the particles -- particularly reactions to INCREASE the particle size -- is not appropriate, especially for in-line tests. Indeed, the fact that Gopinanthan requires particle enlargement indicates that the method of Gopinathan is NOT OPERABLE with particles of native size.

Thus, while the de Boer and present methods need not alter or interfere with the fluid being analyzed, the Gopinathan method causes a reaction in the fluid. The Gopinathan method would have to be carried out by extraction of a sample or it would contaminate the process flow stream.

It is not seen how the Gopinathan disclosure could be combined with the de Boer teaching at all, let alone in a way to arrive Applicant's claimed method --a method that determines a particle size distribution for any number of sizes of particles found in a fluid such as oil; a method that is flexible in that it applies to particles of numerous non-predeteremined sizes and that are found inherently in the oil; and a method that does not change the particles or the oil tested and can even be placed on-line without interfering with the oil composition or flow. Thus, claims 1-19 distinguish patentably over the de Boer reference in view of the Gopinathan patent.

Several of claims 2-19 contain additional features that distinguish even further over de Boer and Gopinathan. For example, Claims 5-9 also call for the steps of the method to be carried out instantaneously. This enables the method to be employed in a near real time manner in a process stream; however, it is hard to imagine how this could be carried out with de Boer, which requires extensive preparation with pre-selected particle sizes, or with Gopinathan, which requires pre-selection of particle sizes, as well as addition of sphere, the carrying out of reactions, and so forth. Claims 7-9 identify particular frequency ranges found to be especially suitable to the present invention and claims 12-14 identify ranges of angles between probe and signal direction. It is not seen how such ranges can be discovered for *any* method not known in the prior art, let alone the methods of the present invention. Claim 15 calls for the resolving of the amplitude versus time data to comprise gating the detected scattered acoustic energy to that part of the detected energy emanating from a focal region and Fourier transforming the amplitude versus time data into a magnitude vs. frequency format. This is nowhere seen or suggested in the art of record and the significance of such steps is clear from the discussion above, wherein the increase in accuracy found to be obtained by resolution such as that incorporating Fourier transformation is discussed. Likewise, the added features of claim 16, which calls for the pulses of acoustic energy to be applied as a tone-burst and the step of resolving of the amplitude versus time data to comprise detecting the magnitude of the scattered energy at selected frequencies, claim 17, which calls for the averaging over the series of pulses the magnitude for each pulse at each selected frequency to produce an average of the magnitude for each selected frequency, and the determination of the agglomerative state of the asphaltenes to be effected by comparing the that average for each selected frequency with a standard, claim 18, which calls for the feature noted with respect to claim 17 wherein the standard is a sample of known particle size, and claim 19, which calls for the additional features of claim 18 wherein the

standard is a model of particle size based on scattering theory, are nowhere seen in the art of record, yet each of such features is an important aspect of the present invention.

Reversal of the rejection of claims 1-11, 15-22 and 24-26 under the judicially created doctrine of obviousness-type double patenting over claims 1-20 of U.S. patent 5,969,237 is respectfully requested. All claims of the present application call for averaging the resolved magnitude versus frequency data. None of the claims of the cited patent refer to any manipulation of the magnitude versus frequency data. As noted above, it has been found that basing the relationship on a single pulse of energy can result in erroneous readings, perhaps due to the orientation of particles at that time of that pulse. The fact that this is not the result of the mere standard averaging out of errors as asserted by the Examiner is shown by the fact that improvement is found by applying a series of pulses, detecting for each of those pulses the back-scattered energy, **resolving the data derived from those detections such as by a Fourier transform** and then averaging the resolved data over the series. If the improved accuracy were the result of mere averaging out errors, one would not expect the improvement associated with first resolving the data such as by a Fourier transform. Claims 15 specifically calls for the resolution to include a Fourier transform --a technique nowhere even hinted at in the prior art of record. Claims 15-19 add other specific features in carrying out the resolution. The claims of the cited U.S. patent nowhere teach or suggest that repeated pulses would do more than simply duplicate the original data, or that averaging the resolved results therefrom would improve the measurement of the agglomerative state. Thus, it is submitted that claims 1-11, 15-22 and 24-26 distinguish patentably over the cited U.S. patent.

Claim 23 has been objected to as depending from a rejected claim, claim 20. In view of the comments above, it is clear that claim 20 should be allowable. Thus, the objection should be withdrawn and action accordingly is respectfully requested.

Thus, it is submitted that all pending claims distinguish patentably over the cited art. In view of the foregoing, favorable reconsideration and early allowance of claims 1-26 are earnestly solicited.

CONCLUSION

In view of the foregoing, it is submitted that all pending claims are allowable. Therefore, it is respectfully submitted that the Examiner be reversed on all outstanding rejections and objections.

Respectfully submitted,

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APPENDIX A

CLAIMS PENDING IN APPLICATION SERIAL NO. 09/509,126

1. (Amended) A method for measuring the agglomerative state of asphaltenes in oil containing asphaltenes, comprising applying to the oil a series of pulses of acoustic energy, each pulse comprising acoustic energy at multiple frequencies, thereby scattering at least part of the energy; detecting, for each of a plurality of pulses in the series, the scattered acoustic energy to produce amplitude versus time data; resolving the amplitude versus time data to obtain a magnitude of the detected scattered acoustic energy at selected frequencies; averaging over the plurality of pulses the magnitude for each pulse at each selected frequency; and determining from the averaging the agglomerative state of the asphaltenes.

2. (Amended) A method as set forth in claim 1 wherein the selected frequencies comprise at least three different frequencies.

3. (Amended) A method as set forth in claim 1 wherein the selected frequencies comprise at least fifteen different frequencies.

4. A method as set forth in claim 1 wherein the steps are carried out without diluting the hydrocarbon liquid.

5. A method as set forth in claim 4 wherein the steps of the method are carried out substantially instantaneously.

6. A method as set forth in claim 5, wherein the detected scattered acoustic energy is back-scattered acoustic energy.

7. (Amended) A method as set forth in claim 6, wherein the scattered acoustic energy is detected over a frequency range of from about 0.1 MHz to about 20 MHz.

8. (Amended) A method as set forth in claim 7, wherein the scattered acoustic energy is detected over a frequency range of from about 0.1 MHz to about 200 MHz.

9. (Amended) A method as set forth in claim 8, wherein the scattered acoustic energy is detected over a frequency range of from about 14 MHz to about 20 MHz.

10. A method as set forth in claim 1, wherein the detecting is carried out by at least one sensor which sensor is incorporated in a signal input probe.

11. A method as set forth in claim 1, wherein the detecting is carried out by at least one sensor which sensor is separate from a signal input probe.

12. A method as set forth in claim 11, wherein the signal input probe and the sensor are located so that the signal direction of the probe intersects the signal direction of the sensor at an angle of less than about 90°.

13. A method as set forth in claim 12, wherein the signal input probe and the sensor are located so that the signal direction of the probe intersects the signal direction of the sensor at an angle of less than about 60°.

14. A method as set forth in claim 13, wherein the signal input probe and the sensor are located so that the signal direction of the probe intersects the signal direction of the sensor at an angle of less than about 45°.

15. (Amended) A method as set forth in claim 1, wherein the resolving of the amplitude versus time data comprises gating the detected scattered acoustic energy to that part of the detected energy emanating from a focal region and Fourier transforming the amplitude versus time data into a magnitude vs. frequency format.

16. (Amended) A method as set forth in claim 1, wherein the pulses of acoustic energy are applied as a tone-burst and the step of resolving of the amplitude versus time data comprises detecting the magnitude of the scattered energy at selected frequencies.

17. (Amended) A method as set forth in claim 1, wherein the averaging over the series of pulses the magnitude for each pulse at each selected frequency produces an average of the magnitude for each selected frequency, and the determining of the agglomerative state of the asphaltenes is effected by comparing the that average for each selected frequency with a standard.

18. A method as set forth in claim 17, wherein the standard is a sample of known particle size.

19. A method as set forth in claim 17, wherein the standard is a model of particle size based on scattering theory.

20. A method as set forth in claim 1, wherein the oil containing asphaltenes is in a process flow stream and the signal of acoustic energy is applied to the oil in the process flow stream.

21. (Amended) A method for measuring the agglomerative state of asphaltenes in an oil containing asphaltenes comprising:

a. removing a sample of the oil and without diluting the oil;

b. applying to the sample a series of pulses of acoustic energy, each pulse comprising acoustic energy at multiple frequencies, thereby scattering at least part of the energy;

c. detecting, for each of a plurality of pulses in the series, the magnitude of the scattered acoustic energy at selected frequencies to produce amplitude versus time data;

d. resolving the amplitude versus time data to obtain a magnitude of the detected scattered acoustic energy at selected incremental frequencies;

e. averaging over the plurality of pulses the magnitude for each pulse at each selected frequency;

f. deriving from the averaging a distribution of the relative size of asphaltene particles scattering acoustic energy; and

g. determining the agglomerative state of the asphaltene particles.

22. A method as in claim 20, having the additional step of returning the undiluted oil sample.

23. A method as in claim 20, wherein the method is carried out in a bench-scale device.

24. (Amended) A method for controlling the agglomeration of asphaltenes in oil which comprises applying a series of pulses of acoustic energy to the oil, each pulse comprising acoustic energy at multiple frequencies, thereby scattering at least a part of the energy; detecting, for each of a plurality of pulses in the series, the scattered energy at selected frequencies to produce amplitude versus time data; resolving the amplitude versus time data to obtain a magnitude of the detected scattered energy at selected incremental frequencies; averaging over the plurality of pulses the magnitude for each pulse at each selected frequency to obtain average magnitude versus frequency data; comparing the average magnitude

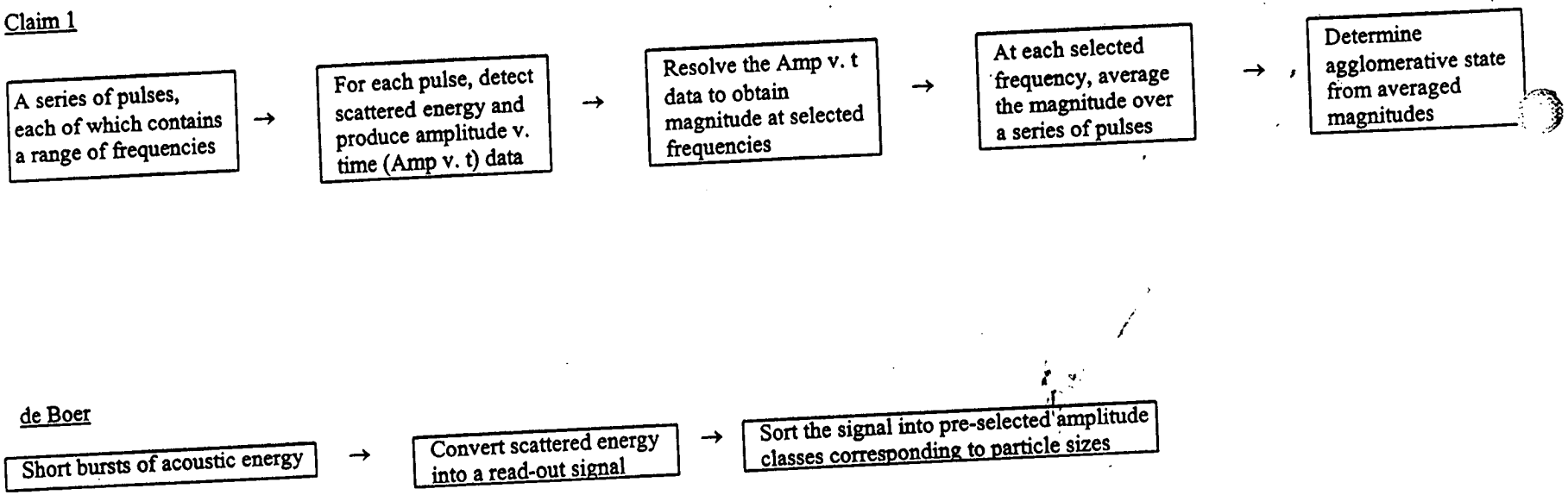
versus frequency data with a standard; and acting to control the number of particles having a particle size corresponding to the selected incremental frequencies.

25. (Amended) A method as set forth in claim 24, wherein the selected frequencies are limited to a frequency range of acoustic energy scattered by the agglomerated asphaltene particles characteristic of the oil.

26. (Amended) A method as set forth in claim 24, wherein the scattered acoustic energy is detected over a frequency range of from about 14 MHz to about 20 MHz.

APPENDIX B

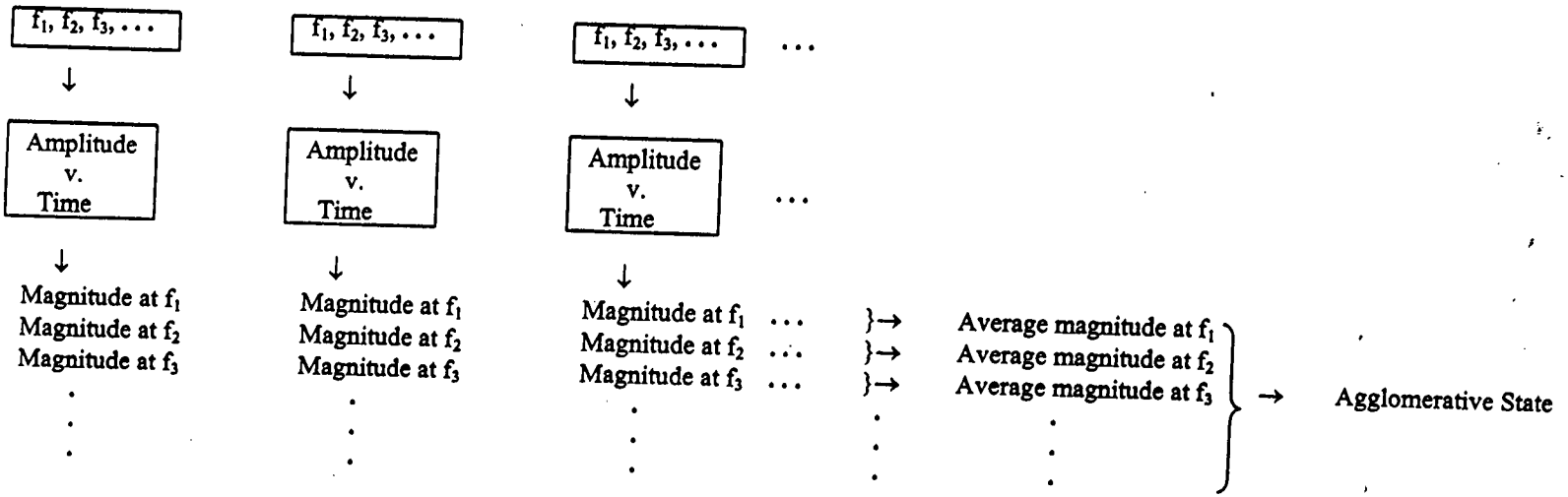
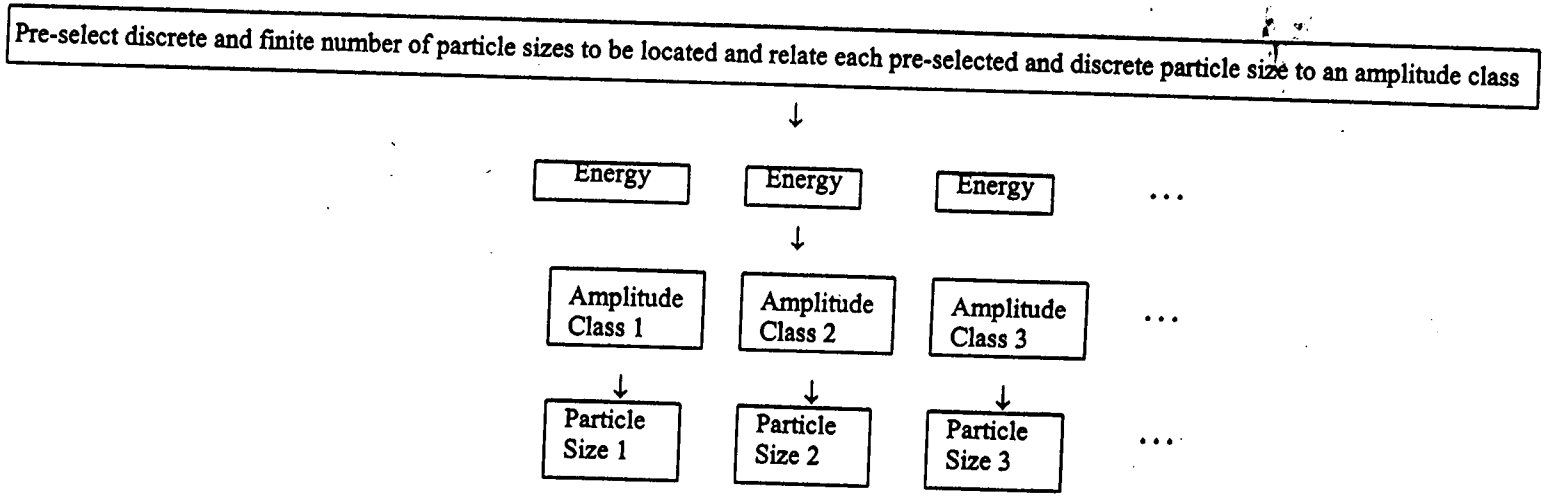
Comparative Flowchart of the Methods of Claim 1 and de Boer



APPENDIX C

Comparative Schematic of the Methods of Claim 1 and de Boer

Claim 1



de Boer